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BURNING RATE TRANSITIONS FOR HMX BURNED AS A BINDERLESS PROPELL--ETC(U)
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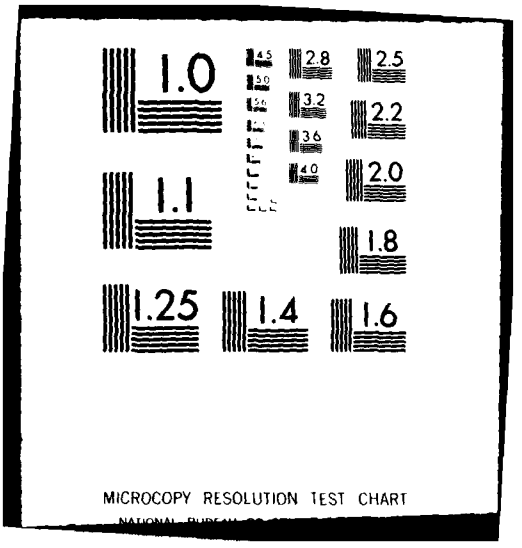
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(6) BURNING RATE TRANSITIONS FOR HMX BURNED AS A BINDERLESS PROPELLANT

JUN 1980

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INTRODUCTION

The burn rates of propellants used in gun and rocket applications can usually be fit to an empirical equation of the form

$$R = AP^n \qquad (1)$$

where R is the burn rate, P is the pressure and A and n are constants. For a typical solid propellant, the pressure exponent n is about 0.8, and the burn rate increases from about 0.5 cm/s at a pressure of 1 MPa (150 psi) to roughly 70 cm/s at 500 MPa (72,500 psi). Rocket motors usually operate at a constant pressure of 69 MPa (10,000 psi) or less. Guns typically operate at peak pressures of 345 MPa (50,000 psi) or higher, although the pressure is much lower than the peak pressure for most of the time the projectile travels down the gun barrel.

Composite (non-homogeneous) propellants containing crystalline HMX (cyclotetramethylene-tetranitramine) or the similar compound RDX (cyclotrimethylene-trinitramine) imbedded in a polymeric binder are gradually replacing homogeneous nitrocellulose-based propellants in many gun and rocket applications. Propellants containing HMX or RDX are commonly called nitramine propellants.

A number of studies have been carried-out to determine the burn rate of pure HMX (1-4), using either large single crystals or pressed powder samples. Single crystal studies have only been successful to about 10 MPa, due to the tendency of the crystals to crack from the thermal stresses associated with burning at pressures higher than this. With pressed powder samples burn rates at pressures up to about

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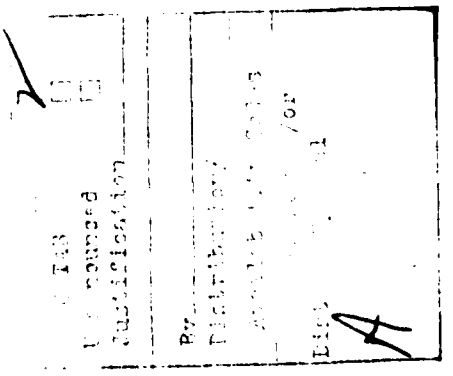
35 MPa (5000 psi) have been successfully measured, and then only by using extremely fine HMX particles (< 5 μm) pressed to very high densities. In most cases it was reported that sample "break up" occurred at a threshold pressure, preventing the burn rate from being measured at higher pressures.

We also have investigated the burn rate behavior of pressed binderless HMX. We have found that the "break up" reported by previous investigators is not random crumbling of the sample but rather an orderly transition from "normal" slow burning to "super-fast" burning. The regression rate increases by roughly three orders of magnitude at the transition pressure, causing the sample to appear to suddenly disappear in a cloud of smoke when observed by conventional photographic techniques. We have developed techniques for measuring very fast regression rates with sufficient accuracy to characterize the super-fast regression beyond the transition point. Using these techniques, we have shown that the regression attains velocities up to 6000 cm/s at high pressure, with pressure exponents as low as 0.3, depending on sample preparation.

These results are significant for certain applications that require extremely rapid gas generation rates. One such application being investigated at this laboratory is the "traveling charge gun" concept. In a traveling charge gun, a single solid cylinder of propellant (or a stack of different propellants) would be attached to the base of the projectile and burn rocket-style as the projectile moved down barrel, burning-out just before muzzle exit. If a near constant pressure were maintained at the propellant (i.e., projectile) base, extremely high ballistic efficiency would result leading to muzzle velocities considerably higher than for a conventional gun cycle for the same maximum gun pressure. Since burning takes place only at the base of the charge, extremely high burn rate propellant (several thousand cm/s or more depending on gun pressure and muzzle velocity desired) is required in order for the gas generation rate to be high enough to maintain constant pressure.

Prior to this study, the only propellant materials known to have burn rates high enough for traveling charge gun operation were certain carborane-based compositions. These materials are very expensive. HMX, on the other hand, is already being produced at low cost for propellant and explosive applications, and its use should make the traveling charge concept economically feasible.



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EXPERIMENTAL

Preparation of HMX Samples

The pressed samples were prepared from three different particle sizes of military grade powdered HMX: as received class E (< 44 μm) and class C HMX screened into two size ranges, 149-297 μm and 105-149 μm. The samples were pressed at 110 MPa (16,000 psi) into parallelepiped strands having the dimensions 3.2 x 0.6 x 0.16-0.19 cm. The density of each parallelepiped was determined from its weight and measured dimensions. The percent theoretical maximum density (TMD), based on a density of 1.90 g/cc for crystalline HMX was 82% for class E, 92% for 105-149 μm class C and 95% for 149-297 μm class C.

One end of each sample was cemented to a holder which mounted securely inside the strand burner (for burn rate measurements) or window chamber (for high-speed photography). To assure one-dimensional (end-to-end) regression during burning, the four large sides of each sample were "inhibited" with a thin coating of epoxy to prevent flame spreading down the sides ahead of the regressing surface.

For the strand burner experiments, the regression rate was determined using two fuse wires -- one passing through the sample near the top, and the other near the base. To accommodate these wires, small (0.35 mm) holes were drilled through the 0.6 x 3.2 cm faces of the sample. The finest size fuse wire that could be handled without breaking (one quarter amp, 0.114 mm diameter) was used in order to achieve as rapid melting as possible. The fuse wire spacing (typically about 2.0 cm) was measured to $\pm$ 0.25 mm (1.5% or better). During an experiment a small current is passed through these wires. Burning of the propellant melts the fuse wires sequentially, generating electrical signals. The regression rate is then determined from the measured time interval and inter-wire distance. The HMX strands were ignited by a small cylindrical piece of JPN propellant that was mechanically attached to the top of the HMX sample and ignited by a heated wire through its center. This technique presumably produced more uniform ignition of the surface.

For the window chamber tests, the samples were not fitted with fuse wires. The samples were directly ignited by a heated wire on the end of the strand to prevent optical interference (flame, smoke, etc.) from burning pieces of JPN propellant.

Strand Burner, Window Chamber and Recording Equipment

In the propellant community, the term "strand burner" has come to mean any reactor in which a single "strand" of propellant can be burned one-dimensionally at constant, or near constant, pressure. Generally, constant pressure is achieved by simply keeping the volume of the system large compared to the mass of sample being burned. (In a "closed bomb", by way of contrast, a number of propellant grains are burned simultaneously on all exposed surfaces, and one measures the self-generated pressure as a function of time).

The strand burner used in these experiments is of conventional design. The main chamber is cylindrical, with a 4.5 cm internal diameter, a 36 cm internal length, and a free volume of 400 cc when assembled with the sample holder. Electrical feed-throughs are provided for the ignition current and time of event (fuse) wires. Compressors are used to prepressurize the burner with nitrogen a few minutes before igniting the sample.

The electrical signals resulting from melting of the fuse wires were recorded on separate channels of a magnetic tape, together with 0.1, 1.0 or 10 ms calibrated time marks. The signals were also used to start and stop a digital counter which measured the corresponding time interval to the nearest microsecond.

The window chamber used for the photographic measurements has a volume of 2700 cc and is equipped with windows sufficiently large to photograph the entire 3.2 cm length of the pressed strands. Movies have been obtained of samples burning at three different pressures: 3.45 MPa (500 psi), 13.8 MPa (2000 psi) and 34.5 MPa (5000 psi). Kodak type 7242 film was used, with a framing rate up to 11,000 frames per second for the fast-burning samples. All movies were obtained with no external illumination of the sample. The camera was started about one second before igniting the sample in order to enable it to reach full speed before sample burning. A blinking light inside the camera was used to put framing rate calibration marks along the edge of the film.

RESULTS

Strand Burner Experiments

The results of the strand burner regression rate measurements for the three particle size pressed HMX samples are shown plotted in Fig. 1.

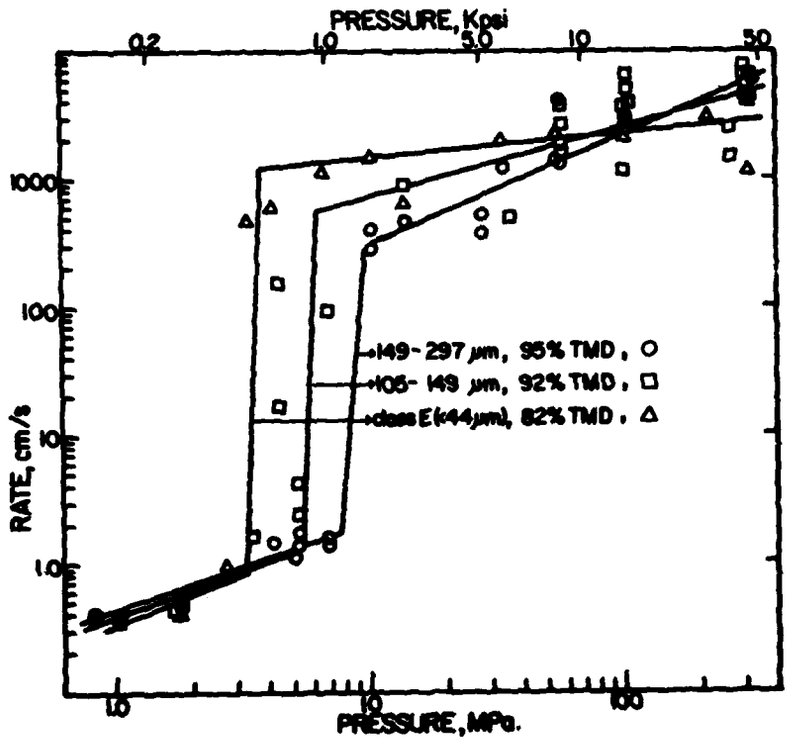

Fig. 1. Regression rate vs pressure for pressed binderless HMX

These samples can be seen to exhibit the following behavior: Below about 3.4 MPa (500 psi), all of the samples exhibit the "normal", slow HMX burn rate measured by other investigators. At higher pressures, each type sample undergoes a transition to super-fast regression. The transition pressure can be seen to increase with increasing particle size and sample density. Over a very small pressure range, the regression rates increase from about 1 cm/s to about 400 to 1000 cm/s. A second break then occurs and the regression rate increases slowly with increasing pressure. At pressures just above the transition pressure, the regression rate is greater, the smaller the HMX particle size. The three curves appear to converge to a rate of about 2500 cm/s at about 100 MPa (14,500 psi), and fall in the approximate range 3000-6000 cm/s at 345 MPa (50,000 psi). In the high pressure region, the apparent burn rate pressure exponent (see Eq. (1)) is less than 0.8 for the coarsest HMX particle size, and less than 0.3 for the finest HMX particle size material.

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The solid lines in Fig. 1 were drawn in by simple visual inspection of the plotted data points. More sophisticated data analysis does not appear to be justified in view of the scatter in the data. As might be expected, reproducibility is worst in the transition and high pressure regions. The most likely explanation for the scatter is the finite melt time characteristics of the fuse wires. For a regression rate of 3000 cm/s and a typical inter-wire distance of 2.0 cm, the expected time interval is only 0.7 ms. For the fuse wires not to contribute to data scatter, they would have to melt on a time scale short compared to this (e.g., 0.1 ms), which is very unlikely. There appears to be much less scatter at high pressure for the class E HMX samples. This will be discussed below in connection with the proposed mechanism.

Window Chamber Experiments

The initial reason for carrying-out the window chamber experiments was to confirm the regression rates measured using the fuse wire technique. For the three pressures investigated (3.45, 13.8 and 34.5 MPa), the regression rates measured photographically agreed with those shown in Fig. 1. Note that at 11,000 frames per second, only 35 frames would record the regression of a 3.2 cm long sample burning down at 1000 cm/s. Much higher framing rate cameras would clearly be required to measure burn rates of 5,000 cm/s or higher.

In addition to confirming the measured strand burner regression rates, the high-speed movies provided valuable clues regarding the mechanism of the super-fast regression above the transition point. At 3.45 MPa (below the transition) the samples were observed to burn slowly with an orange flame, the height of which ($\sim$ 5 mm) is small compared to the length of the sample. At 13.8 or 34.5 MPa (above the transition), the samples burn very rapidly with a white to bluish flame that is very high -- extending beyond the top of the field of view even when the strand has burned down most of the way. (For "normal" burning, flame height would be expected to decrease, not increase, with increasing pressure). Moreover, luminosity fills the volume being observed for some time after the sample has burned down to its base. Finally, it was observed that the regressing surface above the transition pressure is very diffuse and poorly defined. These observations, together with a theoretical analysis, provide the basis for the mechanism proposed in the next section.

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DISCUSSION

Mechanism of the High-Speed Regression

Above a certain transition pressure, the pressed HMX samples studied in this investigation exhibited very high regression rates, and therefore very high mass burning rates, M (g $cm^{-2}s^{-1}$). Mass burning rate can be related to the density of the propellant ρ_s (g/cc), burn rate R (cm/s), burning surface area A (cm^2 per square cm of surface) and regression rate S (cm/s) by

$$M = \rho_s RA = \rho_s S \qquad (2)$$

Consequently, the primary question is whether the transition to high regression rates indicates a dramatic increase in burn rate, or in surface area. That HMX undergoes a transition at low pressures to super-high burn rates is not consistent with the following observations:

1. Other investigators have reported "normal" (slow) burning for their (finer particle size and/or higher density) pressed samples to pressures higher than the transition pressures of our samples.

2. We have burned unconsolidated (loose powder) HMX of different particle sizes in a closed bomb ($P_{max} \sim 190$ MPa). The measured pressurization rates were approximately what would be expected for the estimated surface area and a "normal" burn rate law.

3. The high-speed movies obtained in this study suggest a surface-area-increase mechanism, which is discussed in the following paragraphs.

It follows that the transition to very high mass burn rates is a reflection of an increase in burning surface area. There is more than one way by which this could occur. A number of reports in the literature discuss "convective burning", defined as in-depth burning due to diffusion of hot product gases into the porous surface of the propellant to create a greatly increased burning surface area.

Taylor (5), for example, has studied the burn rate behavior of HMX of different particle sizes, loosely packed to low densities ($\sim$ 1.05 g/cc or 55% TMD) in paper tubes and burned in a strand burner. He showed that these samples underwent a transition toward high apparent burning rate, the transition pressure being greater, the smaller the particle size of the HMX. (This trend is the opposite of that shown in Fig. 1). He interpreted his results in terms of

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convective burning, and concluded that the transition occurs when a molten layer becomes too thin in relation to the pore diameter to provide a barrier to hot gas penetration.

Bobolev, et al. (6), have performed similar studies on low density (1.00-1.04 g/cc) RDX, as well as a number of other propellant materials having widely different melting temperatures. Since all behaved similarly, they concluded that disappearance of a melt layer was not the primary cause of the transition for their low density samples. They proposed instead that convective burning commenced when the pressure reached a critical value relative to the effective diameter of the pores at the surface.

Neither Taylor nor Bobolev attempted to measure regression rates above the transition pressures for their low density samples -- the highest regression rates reported were about 60 cm/s. We have shown that high density pressed HMX attains regression rates up to 3000-6000 cm/s at high pressure. Assuming a normal burning rate law for HMX, this corresponds to an increase in burning surface area of about three orders of magnitude, and is clearly inconsistent with an in-depth convective mechanism -- there is not enough internal surface area near the surface of the sample to explain such an increase.

We therefore propose that the increased surface area does not exist in the solid sample, but is generated by a progressive release of the particles which then burn in a jet flow moving away from the regressing surface, i.e., a "progressive deconsolidation" mechanism operates. In support of this mechanism, we found in our photographic observations a diffuse regression front, a dramatically increased flame height above the transition point, and luminosity persisting for some length of time after the sample has burned down. All these are consistent with such a mechanism for surface area generation. Figure 2 shows an idealized representation of our "progressive deconsolidation" mechanism.

The diffuse regression front results from the fact that there is no sharp boundary between solid and gas at the regressing surface -- particles are continuously breaking loose and accelerating away from the sample. The flame height corresponds to the distance of the particles from the sample when they burn out. The persisting luminosity is simply a reflection of the fact that some of the suspended particles are still burning even though the original sample has long since "disappeared".

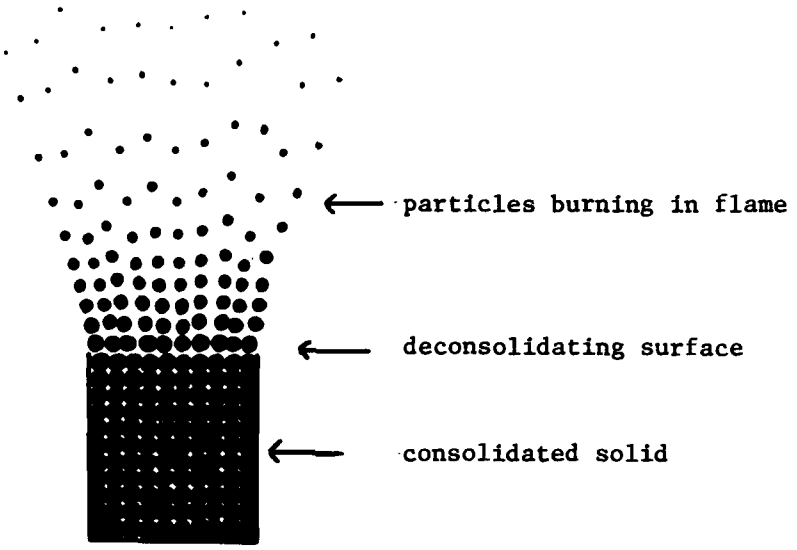


Fig. 2. Idealized representation of progressive deconsolidation mechanism

Theoretical Analysis

We have carried-out modeling calculations to predict the expected flame height (and particle burn-out time) for pressed HMX samples burning at constant pressure by a progressive deconsolidation mechanism. The following simplifying assumptions have been made: a) constant pressure through the flame, b) spherical particles of a single size, c) instantaneous ignition of all particles upon separation from the propellant, d) HMX flame dimensions small compared to the dimensions of the propellant flame, e) no velocity difference between particles and gas, and f) one-dimensional flow, i.e., sample burned in a tube to prevent radial expansion such as shown in Fig. 2.

The equations are as follows. The unit area mass flow rate M (g $cm^{-2}s^{-1}$) is given by Eq. (3) as

$$M = S\rho_s \tag{3}$$

where S is the regression rate and ρ_s is the propellant density. By the law of mass conservation, M is constant at all distances from the

regressing surface (the velocity merely increases when the solid particles convert to gas as they move away from the propellant). For the burn rate R (cm/s) of HMX we take

$$R = 0.264\ P^{0.9} \tag{4}$$

(P in MPa) based on low pressure results in the literature (3). The density of the burned gas ρ_g (g/cc) is determined from

$$1/\rho_g = \frac{I}{P} + b \tag{5}$$

where I is the mass impetus for HMX (1360 Joules/g) and b is the covolume (1.084 cc/g). This equation is based on the Abel-Noble equation-of-state: P(V-b)=RT. The instantaneous density ρ in any region of the two-phase flow is given by

$$\rho = \frac{\rho_c \rho_g}{X\rho_g + (1-x)\rho_c} \tag{6}$$

where ρ_c is the crystalline density of HMX (1.9 g/cc) and X is the mass fraction converted from solid to gas (calculated from the instantaneous sphere volume). From the law of mass conservation, the flow velocity U (cm/s) relative to the regressing surface at any point in the flame is given by

$$U = \frac{M}{\rho} \tag{7}$$

The principal differential equations to be solved involve the rate of change of particle diameter D with respect to time, and with respect to distance from the regressing propellant surface:

$$\frac{dD}{dt} = 2R \tag{8}$$

$$\frac{dD}{dx} = \frac{dD}{dt}\frac{dt}{dx} = \frac{2R}{U} \tag{9}$$

(50)

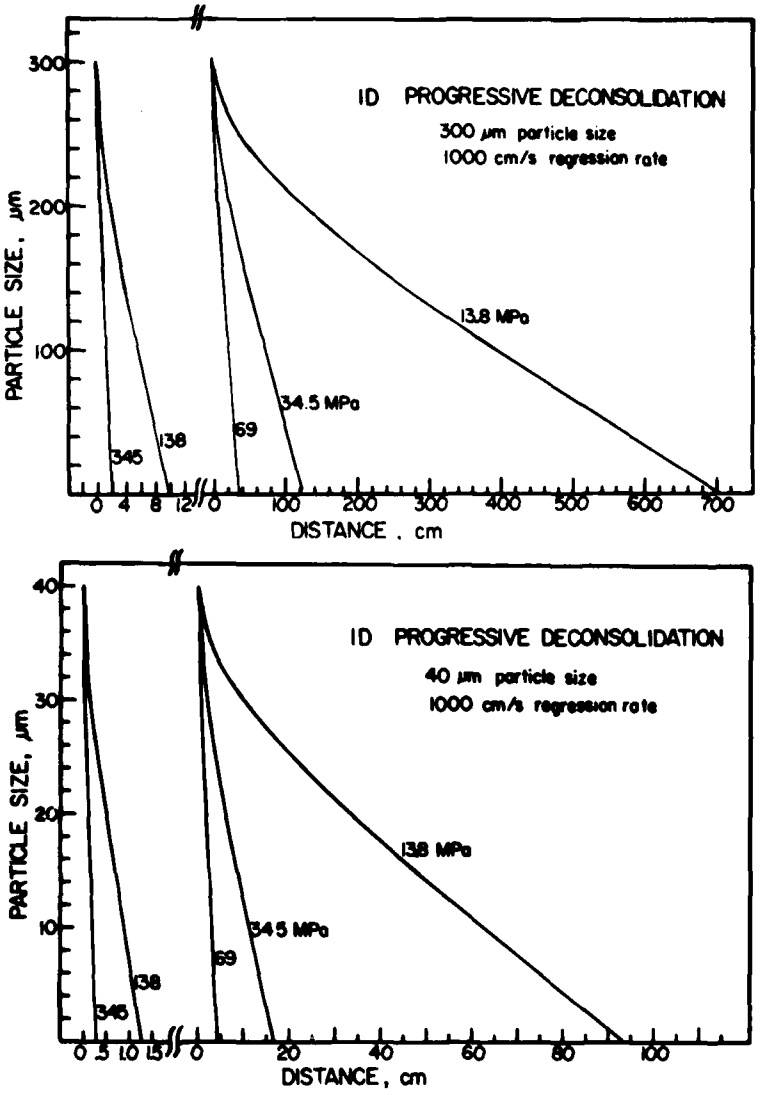
Equation (8) alone is sufficient to calculate the particle burn-out time, and hence the duration of luminescence after the propellant has regressed to its base. For constant pressure, Eq. (8) is a constant, and the calculation is trivial. For example, for 300 micron particles the calculated burn-out time is 5.4 ms at 13.8 MPa, and 2.3 ms at 34.5 MPa. These times are almost as large as the total regression time for the 3.2 cm propellant strands. Actually, luminescence was observed to last for longer than these calculated times, possibly due to agglomeration of some of the particles.

The solution of Eq. (9) in order to determine the burn-out distance (i.e., flame height) is non-trivial, and requires that the instantaneous two-phase density and flow velocity be recalculated using Eqs. (6) and (7), respectively, at each point in the integration. The results of computations at several pressures are shown in Figs. 3 and 4, for 300 μm and 40 μm particles, respectively. A 1000 cm/s regression rate was assumed in these computations. (Computed flame height scales directly with regression rate, as seen in Table I below).

It can be seen that the calculated flame heights are quite large -- a couple of orders of magnitude larger than those expected for "normal" burning where the solid-to-gas conversion occurs at the propellant surface. At the lower pressures (13.8 and 34.5 MPa) where the window chamber photographic experiments were carried out, the calculated flame heights range from 17 to 700 cm, depending on particle size and pressure. Thus, it is not surprising that the top of the flame could not be seen in the photographs. Both the theoretical burn-out times and flame heights for the progressive deconsolidation mechanism are therefore qualitatively consistent with experimental observations.

Note that the calculated flame heights for 300 μm particles are about 7.5 times larger than for 40 μm particles. The greater energy release close to the propellant surface for the finer particles should have led to quicker fuse wire melting in our strand burner experiments. This is the most likely explanation for the greater reproducibility for the class E samples shown in Fig. 1.

The results of this theoretical analysis are shown in Table I, which gives the calculated flame heights, as well as the (gas) density and flow velocity (relative to the regressing surface) for each calculation.



Figs. 3 and 4. Calculated particle size vs distance from propellant for 1D progressive deconsolidation model. Top: 300 μm particle size; Bottom: 40 μm particle size

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Table I. Theoretical Results for One-Dimensional Progressive Deconsolidation

P(MPa)	S(cm/s)	d(cm)	ρ(g/cc)*	U(cm/s)*
		40 μm particles		
344.8	1000	.275	0.198	8,821
137.9	1000	1.32	0.091	19,210
68.95	1000	4.64	0.048	36,520
34.50	1000	16.76	0.025	71,090
13.79	1000	93.91	0.010	175,000
		300 μm particles		
344.8	1000	2.02	0.198	8,821
137.9	1000	9.87	0.091	19,210
68.95	1000	34.76	0.048	36,520
34.50	1000	125.7	0.025	71,090
13.79	1000	704.2	0.010	175,000
344.8	2000	4.05	0.198	17,640
137.9	2000	19.75	0.091	38,410

at burn-out; d is the flame height.

Since the sound speed in gaseous HMX combustion products is about 125,000 cm/s, it can be seen that the flow is supersonic at the lowest pressures. This result is independent of the assumed mechanism since $U = S\ (\rho_s/\rho_g)$ at flame burn-out.

Mechanism of the Transition to High-Speed Regression

Although there is considerable evidence for the progressive deconsolidation mechanism, the reason for the sudden transition to this mechanism at a certain pressure is not obvious. The process responsible for the transition from normal burning to progressive deconsolidation obviously depends on the driving force for the high pressure deconsolidation and this would have to be known before the process responsible for the transition could be understood.

Two possibilities exist to explain the driving force for the deconsolidation process. The first is that one of the normal subsurface processes associated with a burning propellant deconsolidates the charge ahead of the flame front. These include the "thermal wave" (probably too thin compared to particle dimensions to be effective), in-depth absorption of radiation, or a "precursor pressure wave". If one of these subsurface pressure or thermal effects ahead of the

flame front deconsolidate the sample one would expect the mechanical properties of the pressed charge to be important. Since the class C HMX samples are structurally stronger than the class E samples, and since strength increases with density as well, the relative transition pressures for the three samples would not be unexpected.

The second possibility is that burning in the pores very close to the propellant surface breaks the particles loose. This is quantitatively different from in-depth convective burning, and is not inconsistent with the high density of our samples. There is some evidence in the literature for such a mechanism for particle deconsolidation. For example, Andreev, et al. (7), and Belyaev, et al. (8), have both reported transitions from "normal" slow burning toward faster burning for a large variety of pressed propellant and explosive materials. These investigators showed that transition pressure correlates roughly with the permeability of the samples. (Permeability is a measure of the rate of gas flow through a porous sample for a certain pressure difference across the sample). These results suggest that the onset of hot gas penetration into the surface may be responsible for the process of deconsolidation in our samples.

The fact that a wide range of materials exhibit accelerated burning above a critical pressure, together with our results showing that for HMX the resulting super-fast regression is consistent only with a "progressive deconsolidation" mechanism, suggests that progressive deconsolidation may be a general phenomenon. We propose that most and perhaps all materials that have been described as burning by a "convective" mechanism actually burn by progressive deconsolidation, with convection playing a role at most only in the region just below the surface as the possible driving force for the deconsolidation.

CONCLUSIONS, FUTURE WORK

We have shown that pressed high density binderless HMX undergoes a transition to extremely fast regression, which at high pressure attains values of 1000-6000 cm/s with a low pressure exponent. Such regression rates require a greatly increased burning surface area. The mechanism for this appears to be "progressive deconsolidation", rather than "convective" or in-depth burning. This mechanism may be characteristic of all porous propellants which exhibit unusually high apparent burning rates.

A number of additional experimental tests for this mechanism are possible. For example, in a "hybrid" strand burner-closed bomb, pressure would be expected to continue to rise for some time (depending

on particle size) after the sample has burned down, and holography should permit the observation of the particles in the flame.

Although pressed HMX appears to be an attractive, low cost propellant for certain gun applications requiring very high gas generation rates, further characterization is required before this material could be exploited. The mechanical strength of samples prepared in different ways will have to be determined. Since HMX is a "secondary" explosive, the effects of confinement will also have to be investigated. Finally, we will have to determine if samples burn the same way under closed bomb conditions as they do in a constant pressure strand burner. Based on our results to date, pressed HMX looks like a very promising material, and these additional studies would appear to be warranted.

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